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Supporting Information

Ethylenedioxythiophene incorporated diketopyrrolopyrrole conjugated polymers for high-performance organic electrochemical transistors

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Materials and Methods

Materials

Ethylene dioxythiophene (EDOT), 2-ethyrcyano-thiophene, 1-bromo-2-heptyldodecane, dimethyl succinate, *tert*-amyl alcohol, *tert*-butanol potassium, *n*-BuLi, CuCl₂, tris(dibenzylideneacetone)dipalladium(0) (Pd₂(dba)₃), trimethyltin chloride and anhydrous chlorobenzene were purchased from Dickerman Co.. N-Bromosuccinimide (NBS) was used after recrystallization. (3,3'-didodecyl-[2,2'-bithiophene]-5,5'-diyl)-bis(trimethylstannane) was purchased directly from Suntech. Co.. The synthesis routes of monomers and polymers are reported in Supplementary materials.

Materials characterization

The UV-Visible absorption spectra and electrochemical spectra were recorded on PerkinElmer LAMBDA 1050 Spectrophotometer.

Cyclic voltammetry of samples were measured using CHI 660E electrochemical workstation with a standard three-electrode system, with an indium tin dioxide (ITO) coated glass as the working electrode, a platinum wire as the counter electrode and Ag/AgNO₃ (0.01 M in CH₃CN) as the reference electrode. And 0.1 M tetrabutylammonium hexafluorophosphate in acetonitrile (CH₃CN) was used as the supporting electrolyte and ferrocene/ferrocenium (Fc/Fc⁺) as the standard sample. The scanning rate was 50 mV/s. The ionization potential (IP) was estimated by $IP = (E_{ox} - E_{Fc} + 4.8 \text{ V})$.

Electrochemical impedance spectroscopy (EIS) measurements of the polymer films were carried out with a three-electrode system using a Zahner Zennium pro electrochemical workstation, with the platinum wire counter electrode, and Ag/AgCl reference electrode. CPs coated on patterned ITO electrodes (0.2 cm²) acted as the working electrode, and 0.1 M NaCl aqueous solution was

used as the electrolyte. Effective capacitance was calculated from $C \sim 1/(2\pi f Z_{im})$ and used for determination of volumetric capacitance, where f is the frequency and Z_{im} is the imaginary part of the complex impedance.

Nuclear magnetic resonance (NMR) including ^1H NMR (400 MHz) and ^{13}C NMR (100 MHz) spectra were recorded in deuterated solvents on a Bruker ADVANCE 400 NMR Spectrometer. Samples were dissolved in D-chloroform (CDCl_3) with tetramethylsilane (TMS) as the reference of internal standard. The signals have been designated as follows: s (singlet), d (doublet), t (triplet), q (quartet), sd (singlet doublet), dd (doublet doublet) and m (multiplets). J values are expressed in Hz and quoted chemical shifts are in ppm downfield.

Device fabrication and characterization

OECTs were fabricated following a previously reported method.¹ First, chromium/gold source drain electrodes (10/100 nm thick) were deposited and patterned on glass substrates through magnetron sputtering and photolithographic lift off process. The CPs were dissolved in dichlorobenzene at 5 mg/mL and then spin coated on the channel area and annealed at 100 °C for 1 h before patterning. The active channel layer was then patterned between source and drain electrodes through a second lift-off process, forming an active area with 60 μm width, 30 μm length, and 200 ~ 300 nm film thickness. The device was packaged by a layer of SU-8 photoresist to insulate the electrode circuits from the aqueous electrolyte. A platinum wire was used as the gate electrode and 0.1 M NaCl aqueous solution as the electrolyte.

Two Keithley 2400 sourcemeters were employed for the output, transfer and long-term stability characterization of the OECTs, and the data were recorded using a customized LabVIEW program. The gate voltage pulses in the transient measurements were provided by an Agilent 33220A waveform generator and the corresponding device responses were recorded using the Tektronix TBS 2072 digital storage oscilloscope. The synaptic function characterization was performed using Keithley 4200A-SCS semiconductor parameter analyzer.

Synthesis routes

Synthesis of monomers

2,2 3,6-Dithiophen-2-yl-2,5-dihydropyrrolo[3,4-c]pyrrole-1,4-dione (1)²

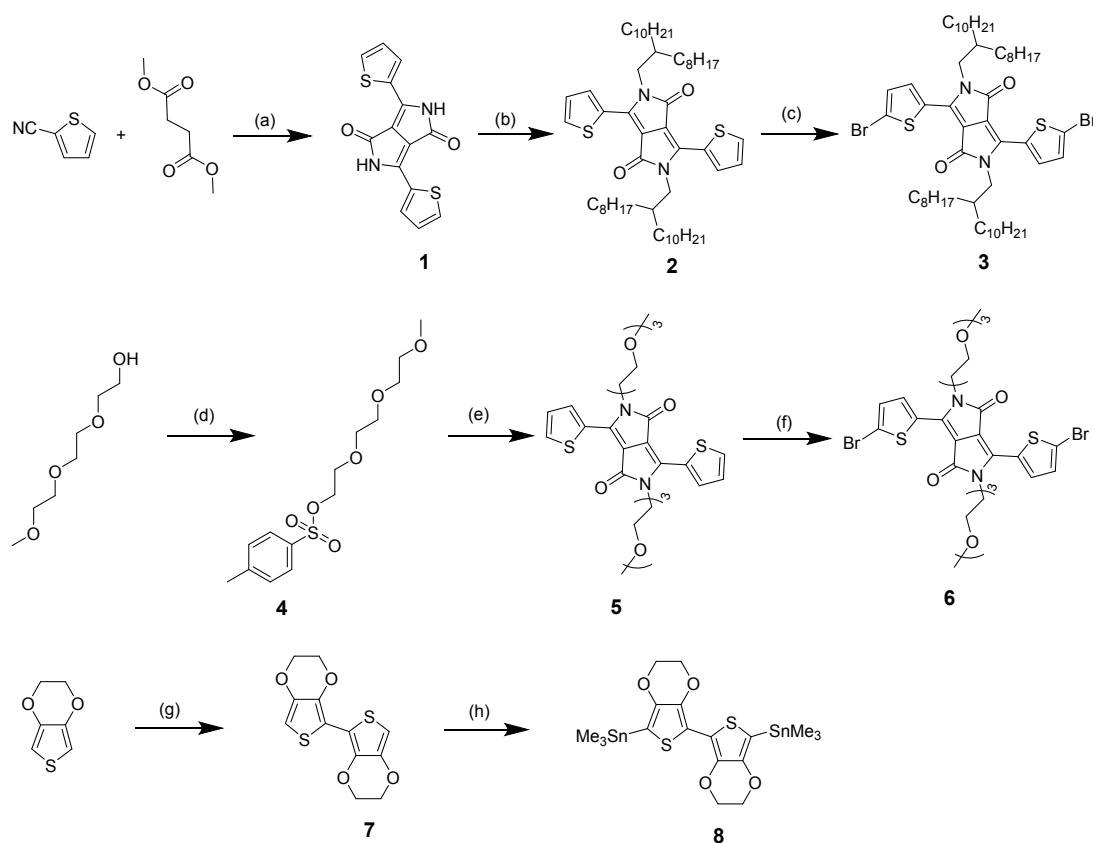
A solution of *tert*-amyl alcohol (75.0 mL) and 2-thiophenecarbonitrile (10.00 g, 109.17 mmol) was injected by a syringe into a round flask with sodium *tert*-butoxide (10.00g, 104.06 mmol) under argon atmosphere. The mixture was warmed to 110 °C, and a solution of dimethyl succinate (3.60 g, 117.8 mmol) in *tert*-amyl alcohol (25 mL) was dropped slowly in 1 h. After the completion of dropping, the reaction was kept at the same temperature for about 1 h, then the byproduct of methanol was distilled off and the reaction was kept for another 2 h. Then the mixture was cooled to 65 °C, diluted with 50 mL of methanol, and neutralized with acetic acid and refluxed for another 10 min. Then the suspension is filtered, and the black filter cake is washed by hot methanol and water twice and dried in vacuum to afford coarse product and could be used directly for next step without further purification (6.3 g, yield 85%).

2,5-Bis(2-octyldodecyl)-3,6-di(thiophen-2-yl)-2,5-dihydropyrrolo[3,4-c]pyrrole-1,4-dione (2)³

Compound **1** (13.00 g, 43.30 mmol) and anhydrous potassium carbonate (24.0 g, 173.0 mmol) dissolved in DMF (250.0 mL) in a two-neck round flask were heated to 145 °C under N₂ atmosphere. 2-Octyldodecyl bromide (38.60 g, 200.0 mmol) was added one portion by syringe. When the reaction was further stirred for 15 h at the same temperature, the solution was cooled to room temperature, and poured into 500 mL of ice-water, and then filtered. The filter cake was washed several times with water and methanol. After drying under vacuum, the crude product was purified by silica gel chromatography using dichloromethane as eluent and a purple-black solid powder was collected. (17.30 g, yield 76%). ¹H NMR (CDCl₃, 400 MHz): δ 8.95 (d, 2H), 7.62 (d, 2H), 7.27 (d, 2H), 4.03 (m, 4H), 1.85 (m, 2H), 1.36-1.22 (m, 16H), 0.85 (m, 12H) ppm.

3,6-Bis(5-bromothiophen-2-yl)-2,5-bis(2-octyldodecyl)-2,5-dihydropyrrolo[3,4-c]pyrrole-1,4-dione (3)

Compound **2** (1.0 g, 1.16 mmol) and N-bromosuccinimide (0.44 g, 2.5 mmol) were dissolved into chloroform (30.0 mL) in a two-neck round flask under argon atmosphere, and then the solution was protected from light and stirred at room temperature for 24 h. Then the mixture was poured into 50 mL of methanol and then filtered. The filter cake was washed with hot methanol twice. After drying in vacuum, the pure product was obtained as a purple-black solid (0.96g, yield 81%).
¹H NMR (CDCl₃, 400 MHz): δ 8.64 (d, 2H), 7.18 (d, 2H), 3.91 (m, 4H), 1.82 (m, 2H), 1.36-1.24 (m, 16H), 0.86 (m, 12H) ppm.



Scheme S1. Reactions conditions: (a) sodium *tert*-butoxide, *tert*-amyl alcohol, 110 °C. (b) octyldodecyl bromide, anhydrous K₂CO₃, DMF, reflux. (c) NBS, AcOH/CHCl₃, 0 °C; (d) TsOCl, NaOH, THF/H₂O, 0 °C. (e) anhydrous K₂CO₃, DMF/H₂O, reflux. (f) NBS, AcOH/CHCl₃, 0 °C. (g) *n*-BuLi, -78 °C; CuCl₂, THF, 0 °C. (h) *n*-BuLi, Me₃SnCl, THF, -78 °C.

2-(2-(2-Methoxyethoxy)ethoxy)ethyl-4-methylbenzenesulfonate (TsO-TEG, 4)⁴

Triethylene glycol monomethyl ether monotosylate (TEG) was prepared according to methods in literature. A solution of triethylene glycol monomethyl ether (6.0 g, 36.54 mmol) in THF (35.0 mL) was cooled to 0 °C. Then 35 mL aqueous solution of NaOH (7.0 g, 175mmol) was added dropwise over 5mins, followed by dropwise addition of 40.0 mL Tosyl chloride (TsOCl) (8.36 g, 1.2eq) in THF within 2 h. The reaction mixture was stirred for another one hour at 0 °C, then left to warm to room temperature (RT), and further stirred for 12 h. The resulting solution was poured into aqueous hydrochloric acid and extracted with DCM. The organic phase was separated and washed three times with 50 mL water each, then dried over anhydrous MgSO₄, and concentrated in vacuum to give **4** as a colorless liquid. Yield: 11.0 g (34.00 mmol, 95%). ¹H NMR (400 MHz, CDCl₃): δ 2.43 (s, 3H); 3.35(s, 3H,); 3.51(m, 2H); 3.56-3.60 (m, 6H); 3.67(t, *J* = 4.9Hz,2H); 3.15(t, *J* = 4.9Hz,2H); 3.32(d, *J* = 8.2Hz,2H); 7.78(d, *J* = 8.2Hz,2H) ppm.

2,5-Bis(2-(2-(2-methoxyethoxy)ethoxy)ethyl)-3,6-di(thiophen-2-yl)-2,5-dihydropyrrolo[3,4-c]pyrrole-1,4-dione (5)⁵

A solution of compound **1** (1.50 g, 5.00 mmol) and anhydrous potassium carbonate (4.20 g, 30.0 mmol) in DMF (50.0 mL) in a two-neck round flask was heated to 120 °C under N₂ atmosphere. TsO-TEG (**4**) (6.36 g, 20.0 mmol) was added one portion by syringe. After stirring overnight at the same temperature, the solution was cooled to room temperature and poured into 200 mL of ice-water, then filtered. The filter cake was washed with water and methanol several times. After drying in vacuum, the crude product was purified on silica gel chromatography using DCM: hexane: ethyl acetate = 2:2:1 as eluent to give a purple-black solid. (1.60 g, yield 55%). ¹H NMR (CDCl₃, 400 MHz): δ 8.76 (d, 2H), 7.64 (d, 2H), 7.27 (d, 2H), 4.28 (m, 4H), 3.79(m, 4H), 3.65-3.45 (m, 16H), 3.35(s, 6H) ppm.

3,6-Bis(5-bromothiophen-2-yl)-2,5-bis(2-(2-(2-methoxyethoxy)ethoxy)ethyl)-2,5-dihydropyrrolo[3,4-c]pyrrole-1,4-dione (6)⁶

A solution of 2,5-bis{2-[2-(2-methoxyethoxy)ethoxy]ethyl}-3,6-di(thiophen-2-yl)pyrrolo[3,4-c]pyrrole-1,4(2H,5H)-dione (1.0 g, 1.68 mmol) in dry chloroform (30.0 mL) in a round-bottom flask covered with aluminium foil was cooled to 0 °C and added with N-bromosuccinimide (0.61 g, 3.40 mmol) in portions. The reaction mixture was stirred at 0 °C in the dark for 6 h, then quenched with a saturated aqueous solution of NH₄Cl (30.0 mL), and then extracted with DCM (3×60 mL). The organic phase was washed with an aqueous brine water (3×30 mL), dried with MgSO₄ and concentrated under vacuum. The crude product was purified on plate chromatography with DCM:methanol (5:1) as eluent. Red needle crystals were obtained after recrystallization from dichloromethane and hexanes. After drying in vacuo, pure product 6 was obtained as a dark Red solid (0.697 g, 55 % yield). ¹H NMR (400 MHz, CDCl₃): δ 8.44 (d, *J* = 4.0 Hz, 2 H), 7.14 (d, *J* = 4.0 Hz, 2 H), 4.11 (t, *J* = 5.8 Hz, 4 H), 3.71 (t, *J* = 5.8 Hz, 4 H), 3.61-3.56 (m, 4 H), 3.55-3.49 (m, 8 H), 3.46-3.41 (m, 4 H), 3.29(s, 6H) ppm.

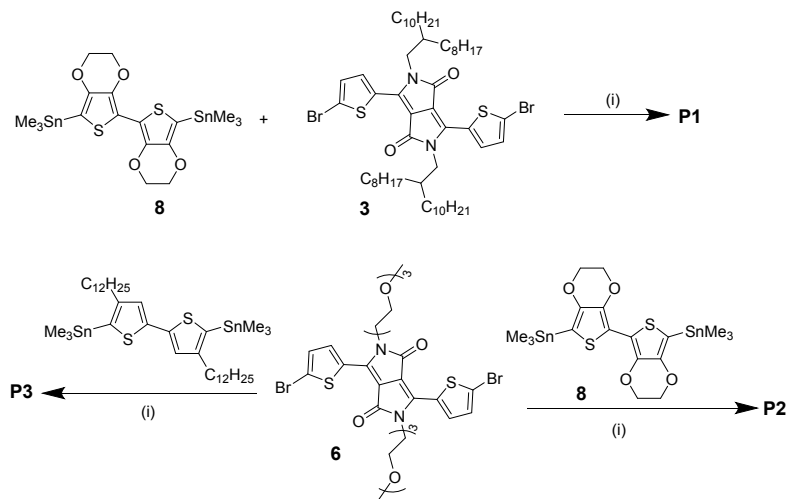
2,2',3,3'-Tetrahydro-5,5'-bithieno[3,4-b][1,4]dioxine (7)⁷

In a 250 ml two-necked flask under N₂ atmosphere, EDOT (8.0 g, 56.3 mmol) in 160 mL dry THF was cooled to -78 °C and added dropwise with 23.2mL *n*-BuLi (2.5 M in hexane). The reaction solution was restored to 0 °C and kept for 30 min. Next, anhydrous CuCl₂ (8.0 g, 59.5 mmol) was added and stirred overnight. The reaction solution was poured into crushed ice and extracted with DCM (3×50 ml). The crude product was passed through a packed diatomite, and the liquid was evaporated to dry under vacuum and afforded 3.2 g product 7. ¹H NMR (CDCl₃): δ 6.27 (s, 2H), 4.24–4.33 (m, 8H) ppm.

7,7'-Bis(trimethylstannyl)-2,2',3,3'-tetrahydro-5,5'-bithieno[3,4-b][1,4]dioxine (8)⁸

Compound 7 (0.85 g, 3.0 mmol) in dry THF (30.0 ml) was cooled to -78 °C, followed with dropwise addition of *n*-BuLi (2.5 mL, 2.5 M in hexanes, 6.15 mmol) within 2 h. Then trimethyltin chloride (9.0 ml, 1 M in hexane, 9.00 mmol) was added at -78 °C. The reaction solution was slowly warmed to room temperature and kept stirring overnight under nitrogen atmosphere. The solvent

was removed and the solid was dissolved in CH_2Cl_2 (50.0 ml), washed with water (300 ml), dried over anhydrous Na_2SO_4 , filtered and evaporated to dryness to give a brown solid. Further recrystallized from hexane gave 0.90 g compound **8** as a beige solid (50%). $^1\text{H NMR}(\text{CDCl}_3)$: δ 4.25-4.35 (t, 4H), 4.22-4.19 (t, 4H) ppm.



Reaction conditions: (i) $\text{Pd}_2(\text{dba})_3$, $\text{P}(o\text{-tolyl})_3$, chlorobenzene, reflux.

Synthesis of polymers^{5,6}

P1 In the glove box with N_2 atmosphere, compounds **8** (76.9 mg, 0.13 mmol), **3** (95.0 mg, 1 equivalent), 3% equivalent $\text{Pd}_2(\text{dba})_3$ and 12% equivalent tris(2-methylphenyl)phosphine ($\text{P}(o\text{-tolyl})_3$) and 4 ml anhydrous chlorobenzene were packed in Schlenk tube with stirring magneton and then slowly heated to 120 °C in an oil bath for 80 h continuous reaction. After restored to room temperature, the reaction solution was poured into a mixture of HCl:methanol (15 mL:200 mL) and further stirred for 24 h before filtration. Crude polymer product was purified in a Soxhlet extractor with acetone, methanol, DCM solvent, respectively, and then chloroform and chlorobenzene. The deep colored solution was evaporated to dry and continued to dry in under vacuum at 100 °C for 12 h, affording solid blue membrane as **P1**.

P2 Following the synthetic procedure for P1 using compound **6** instead of **3**, polymer **P2** was prepared similarly as a solid blue membrane.

P3 Following the similar procedure as P1 and P2, the Stille coupling polymerization between Bis(3,3'-didodecyl-[2,2'-bithiophene]-5,5'-diyl)bis(trimethylstannane) (93.8 mg, 0.11 mmol) and compound **6** (85.0 mg, 1 equivalent) gave **P3** as a solid blue membrane.

Table S1. The gel-permeation chromatography (GPC) and differential scanning calorimetry (DSC) data of P1, P2, and P3.

| Samples | T _g (°C) | T _c (°C) | T _m (°C) | T _d (°C) | M _n (Da) | M _w (Da) | PDI |
|-----------|---------------------|---------------------|---------------------|---------------------|----------------------|---------------------|------|
| P1 | – | 295 | 305 | 383 | 8.63×10 ⁴ | 1.2×10 ⁵ | 1.42 |
| P2 | – | 217 | 300 | 365 | 7.80×10 ⁴ | 9.7×10 ⁴ | 1.25 |
| P3 | – | 302 | 332 | 390 | 1.18×10 ⁵ | 1.9×10 ⁵ | 1.65 |

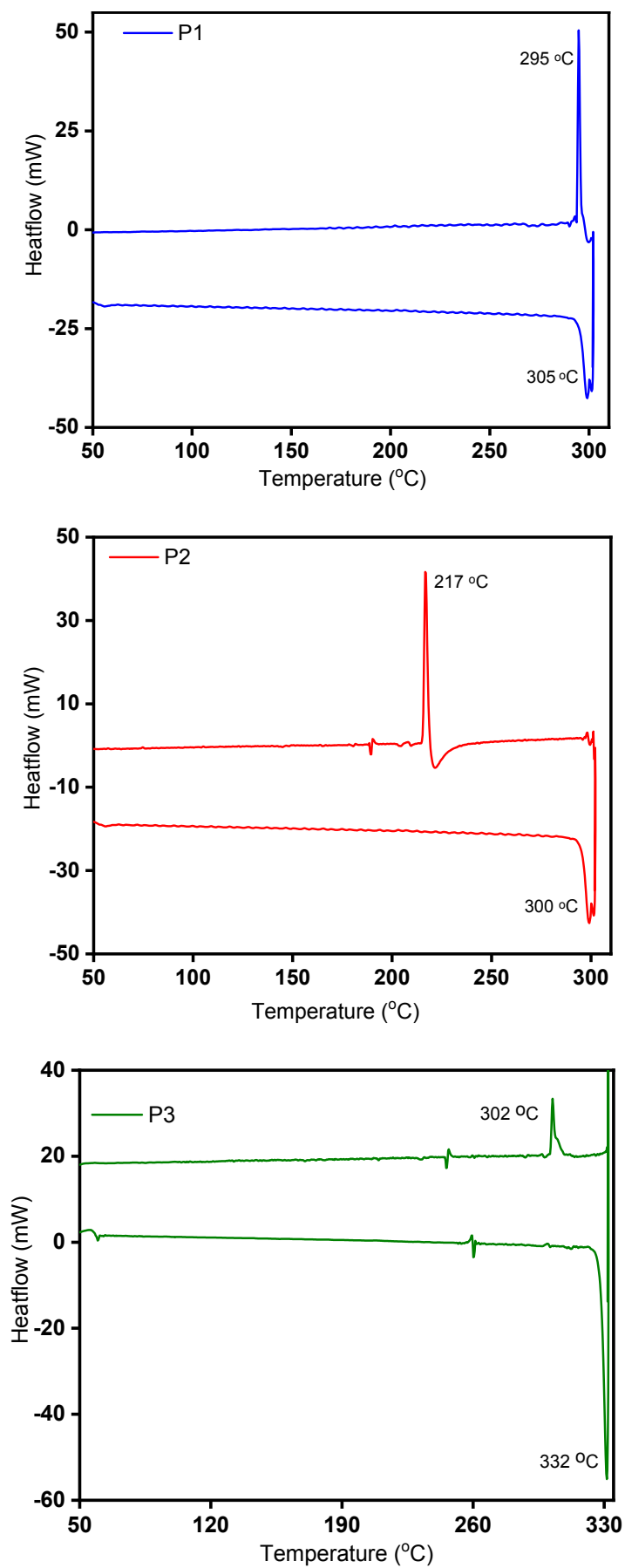


Figure S1. The differential scanning calorimetry (DSC) curves of P1, P2, and P3.

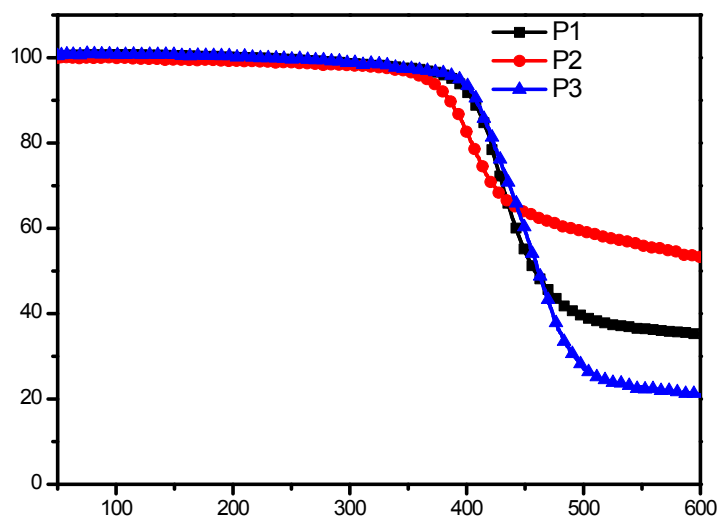


Figure S2. The thermogravimetric Analysis (TGA) spectra of P1, P2, and P3.

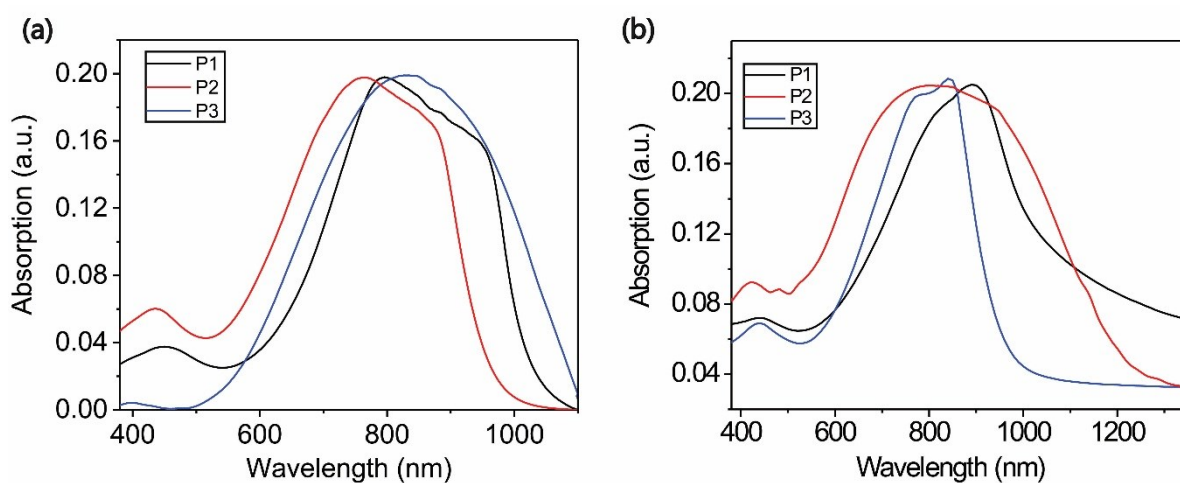


Figure S3. UV-vis absorption spectra of P1, P2 and P3 in (a) CHCl_3 (1×10^{-5} g/ml) and (b) films.

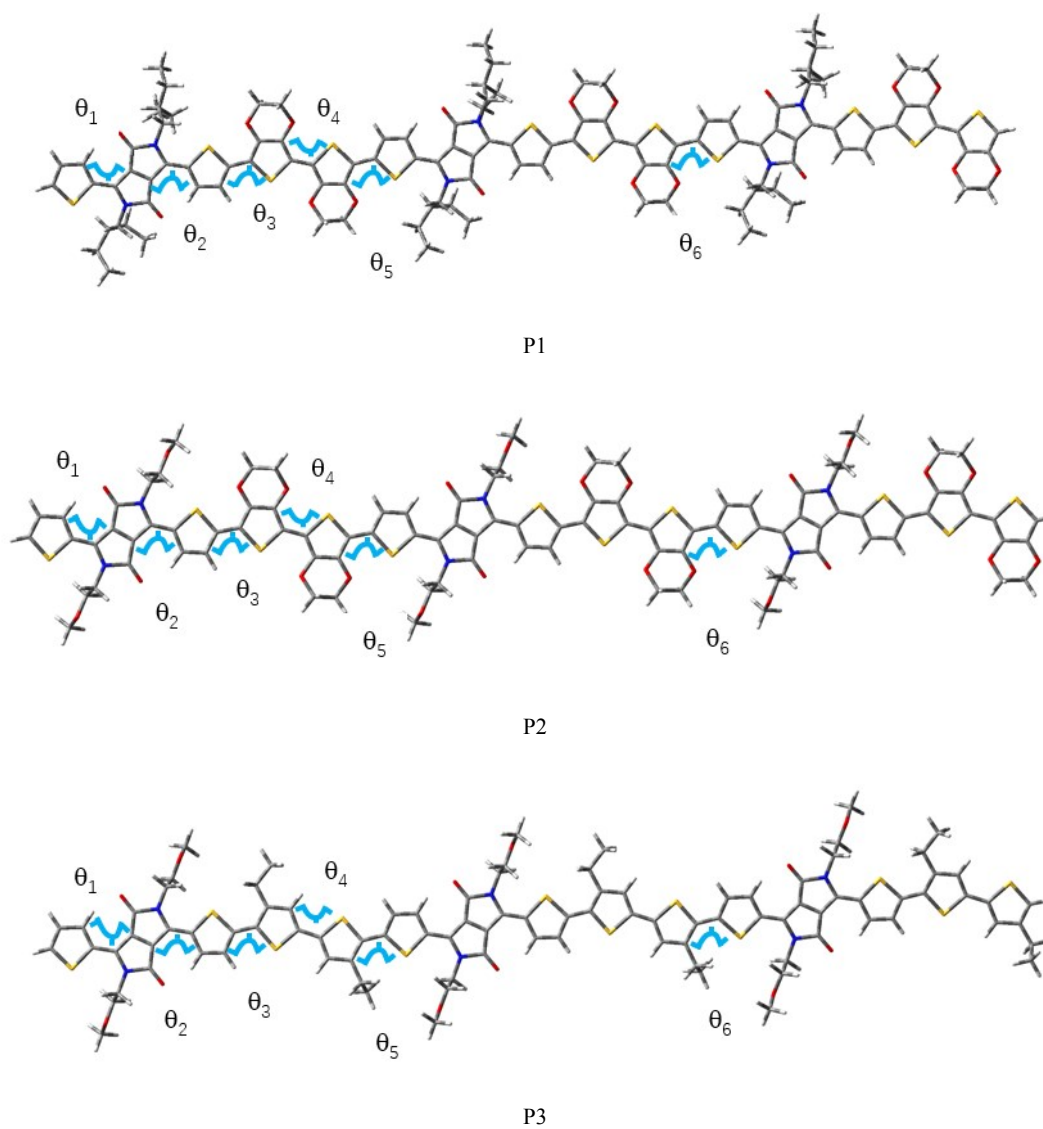


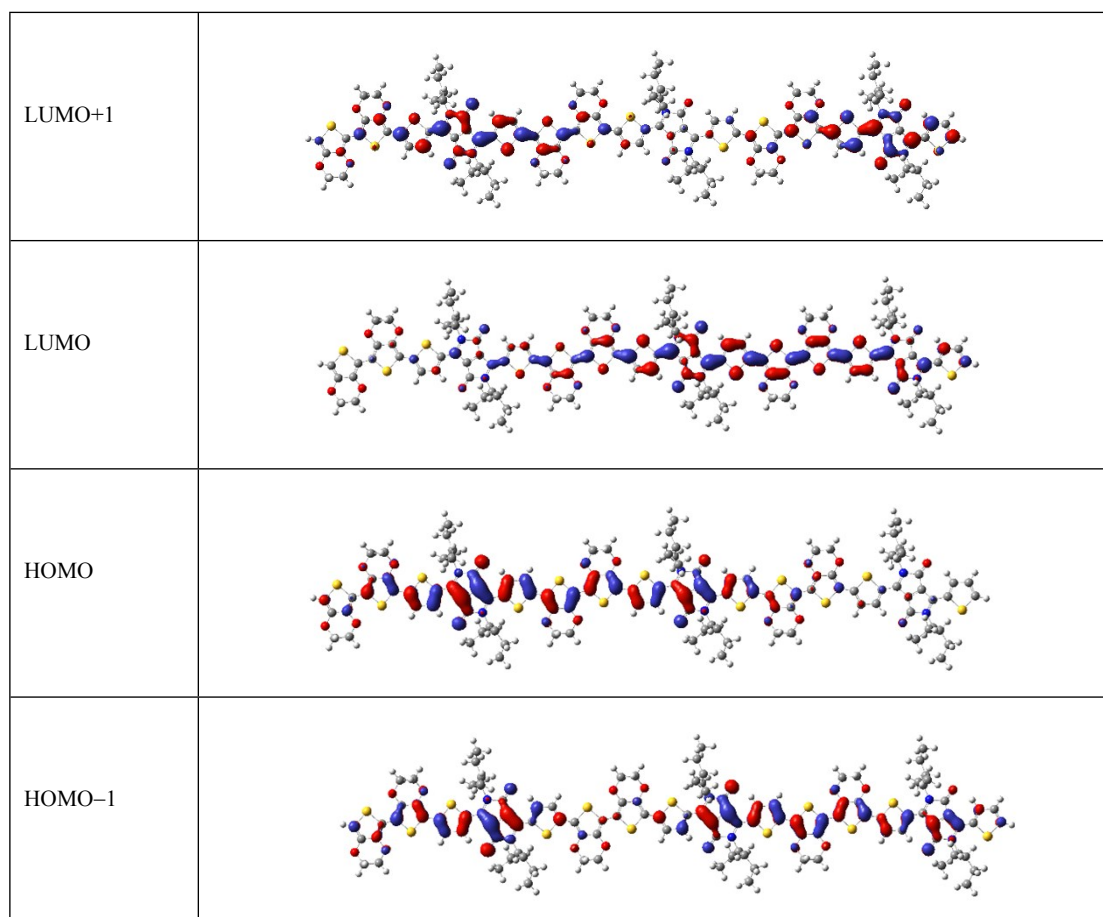
Figure S4. Ground state geometries of the polymers P1, P2 and P3.

Table S2. Inter-ring Angle of trimers of aDTDPP-bis-EDOT, gDTDPP-bis-EDOT, and gDTDPP-aBT.

| Angle / ° | θ_1 | θ_2 | θ_3 | θ_4 | θ_5 | θ_6 |
|-----------|------------|------------|------------|------------|------------|------------|
| P1 | 2.65 | -1.18 | -0.09 | -0.68 | -1.01 | -0.66 |
| P2 | 0.09 | -0.54 | -0.22 | 1.05 | 0.61 | 1.16 |
| P3 | -0.45 | -0.19 | -0.68 | -1.61 | 8.03 | 8.86 |

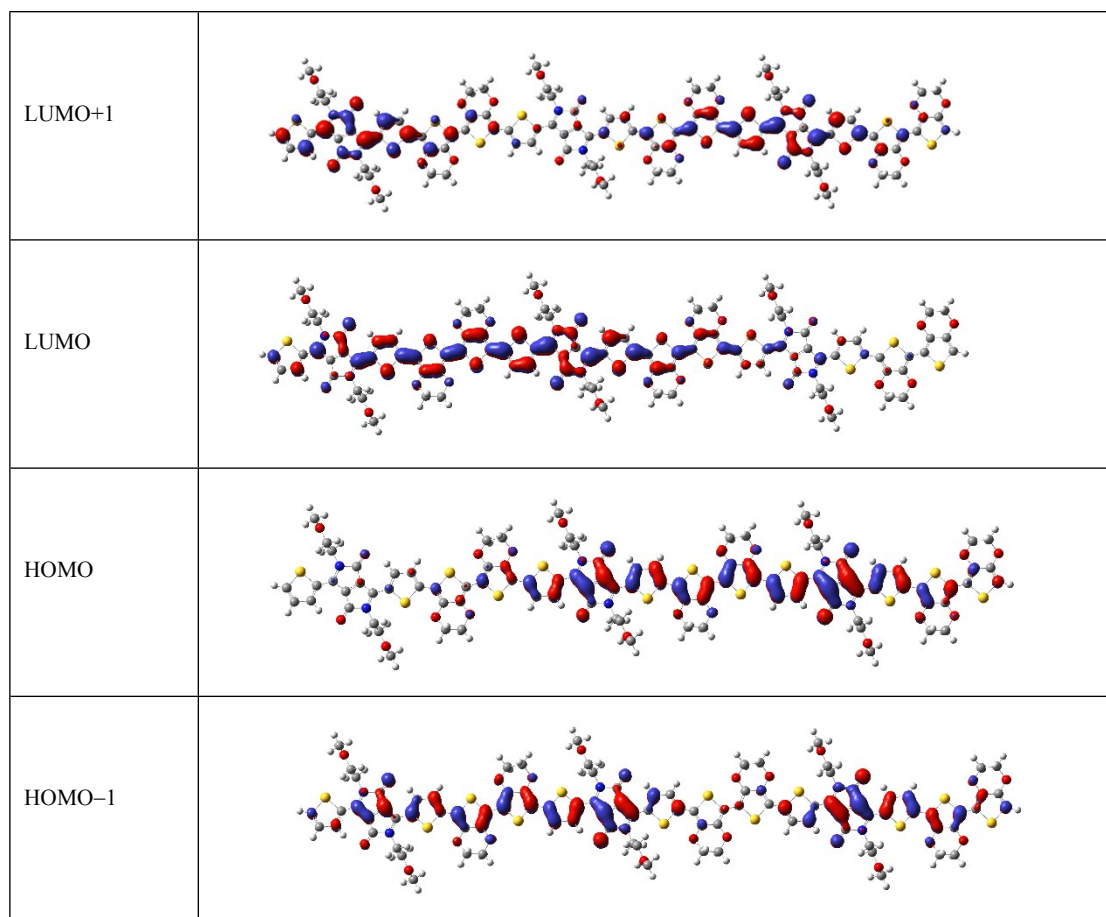
Table S3. Frontier molecular orbital energy levels of P1, P2 and P3.

| MO/ eV | HOMO-1 | HOMO | LUMO | LUMO+1 |
|--------|--------|-------|-------|--------|
| P1 | -4.53 | -4.33 | -2.85 | -2.64 |
| P2 | -4.53 | -4.33 | -2.85 | -2.63 |
| P3 | -4.86 | -4.70 | -3.12 | -2.91 |



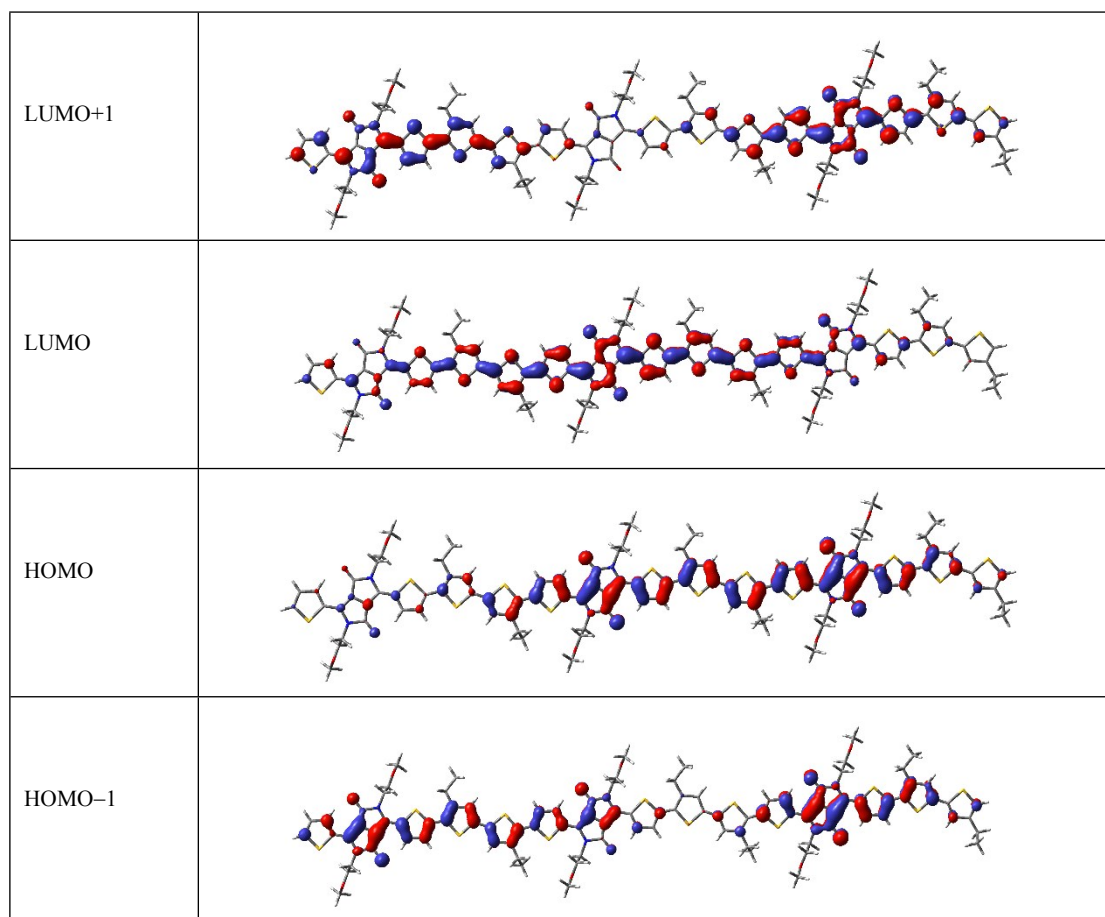
MO = 0.02, Density = 0.0004.

Figure S5. Electron density distribution of Frontier molecular orbitals of P1.



MO = 0.02, Density = 0.0004.

Figure S6. Electron density distribution of Frontier molecular orbitals of P2.



MO = 0.02, Density = 0.0004.

Figure S7. Electron density distribution of Frontier molecular orbitals of P3.

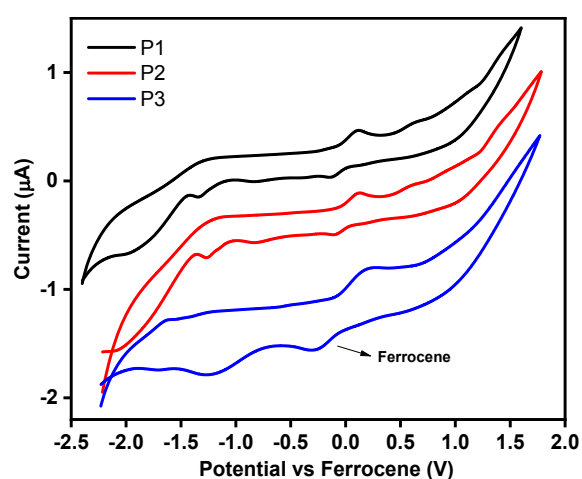


Figure S8. Cyclic voltammograms of the polymers recorded in CHCl_3 solution (1×10^{-4} M) with 0.1 M Bu_4NPF_6 as electrolyte (working electrode: glassy carbon; counter electrode: Pt wire; reference electrode: non-aqueous Ag/Ag^+ ; ferrocene was used as internal standard for potential calibration).

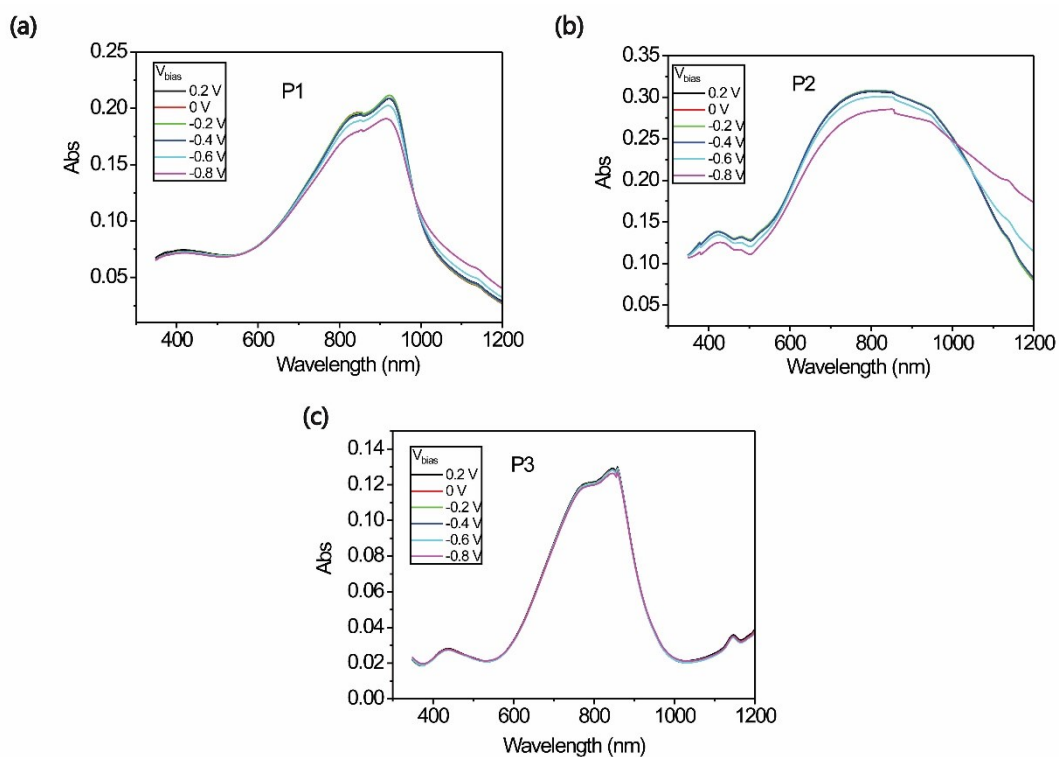


Figure S9. UV-Vis Spectroelectrochemistry of thin films of (a) P1, (b) P2 and (c) P3 in 0.1M NaCl aqueous solution. The films are coated on ITO substrates.

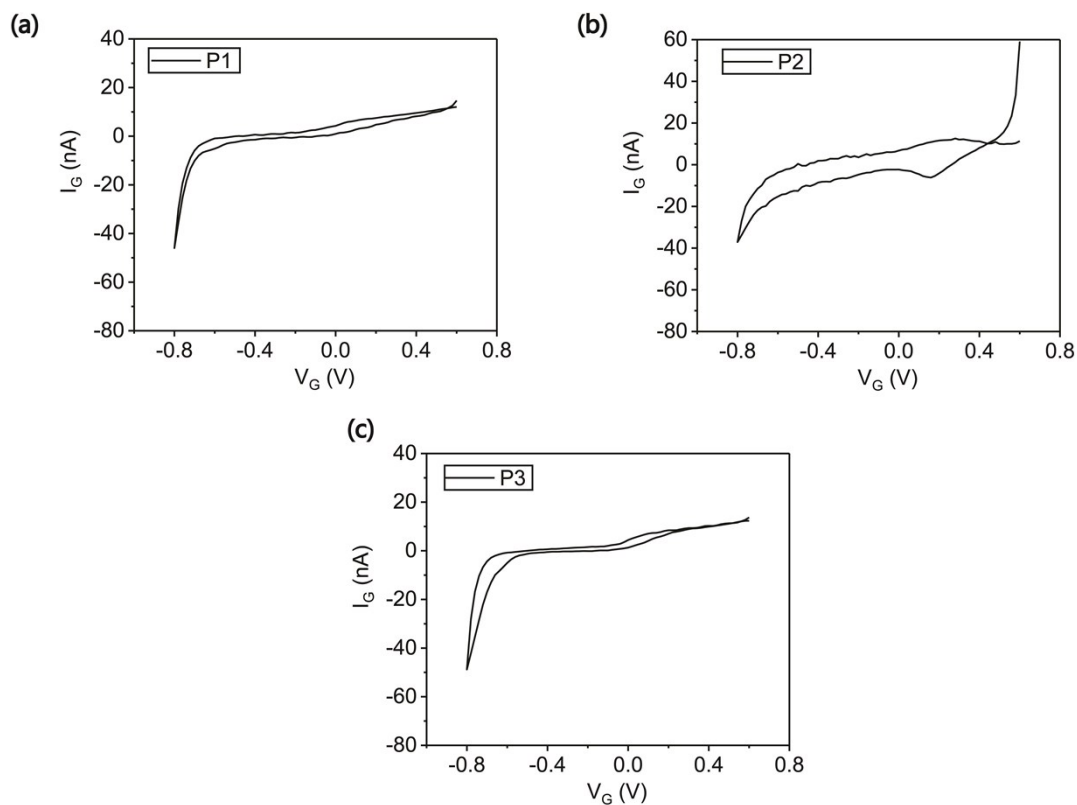


Figure S10 gate leakage current of (a) P1, (b) P2 and (c) P3 based OECT corresponding to the transfer characterization in Figure 2.

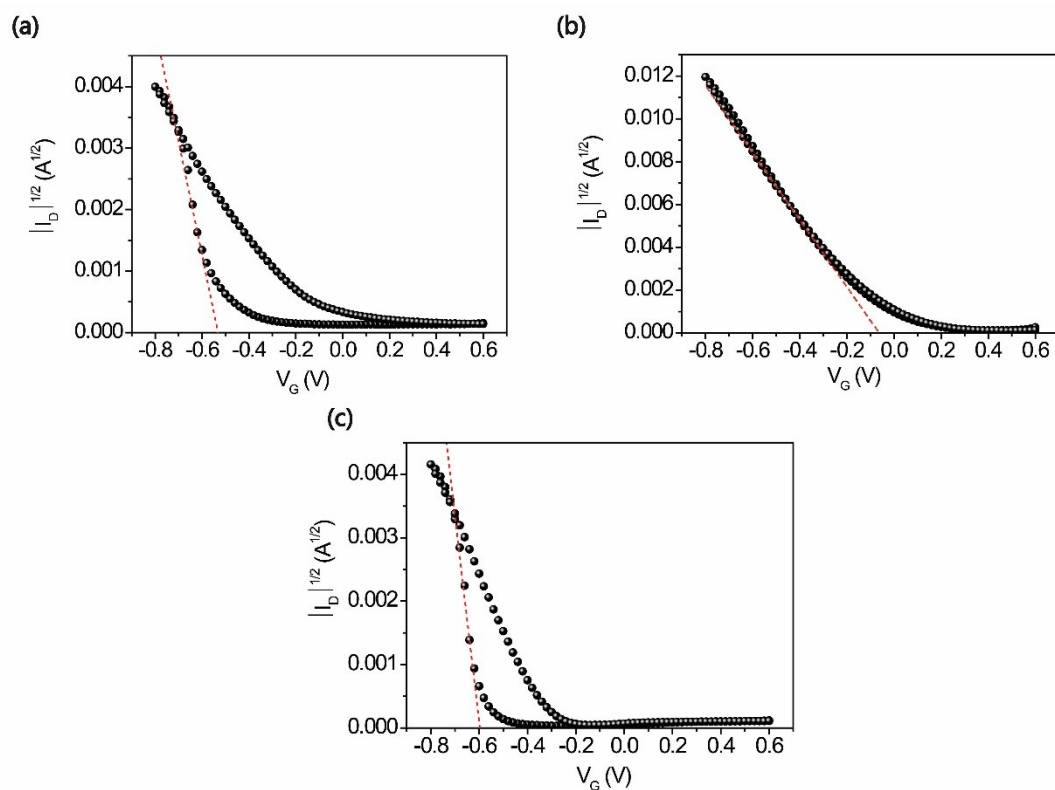


Figure S11. Determination of threshold voltage. Square root of the absolute value of channel current as a function of V_G for (a) P1, (b) P2 and (c) P3 based OECTs. The crosspoint of the tangent in the forward scan curves (red dash line) with the x-axis was determined as the threshold voltage of the device.

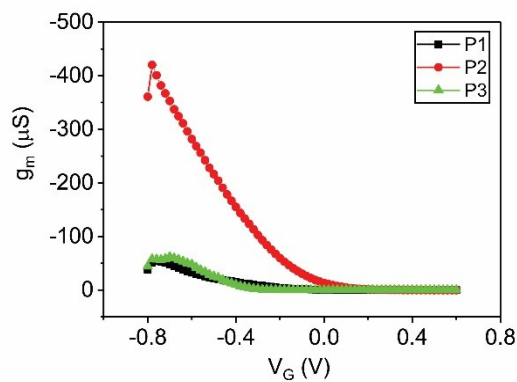


Figure S12. Gate voltage (V_G) dependence of transconductance for OECTs based on P1, P2 and P3 polymers. Drain voltage $V_D = -0.6$ V.

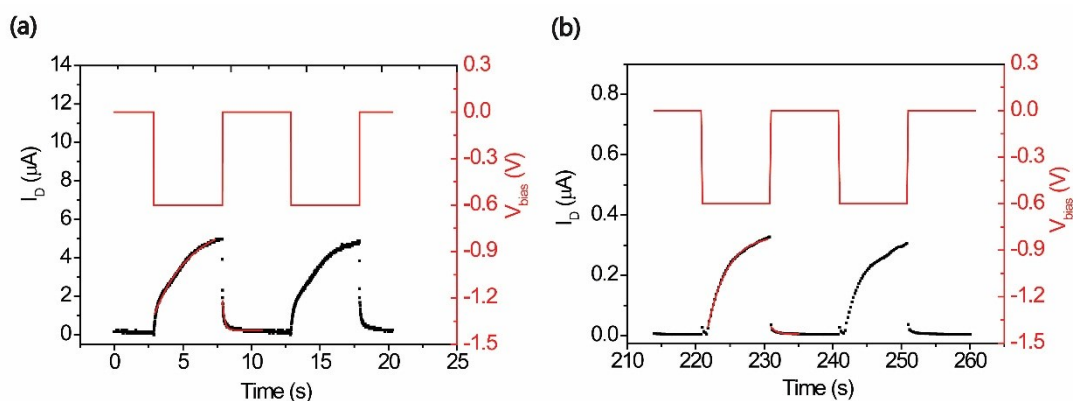


Figure S13. Transient current response (left y-axis) of (a) P1 based OEET and (b) P3 based OEET corresponding to the -0.6 V pulsed gate voltage applied (right y-axis). The fitting lines stands for the exponential decay fits for the charging/discharging processes.

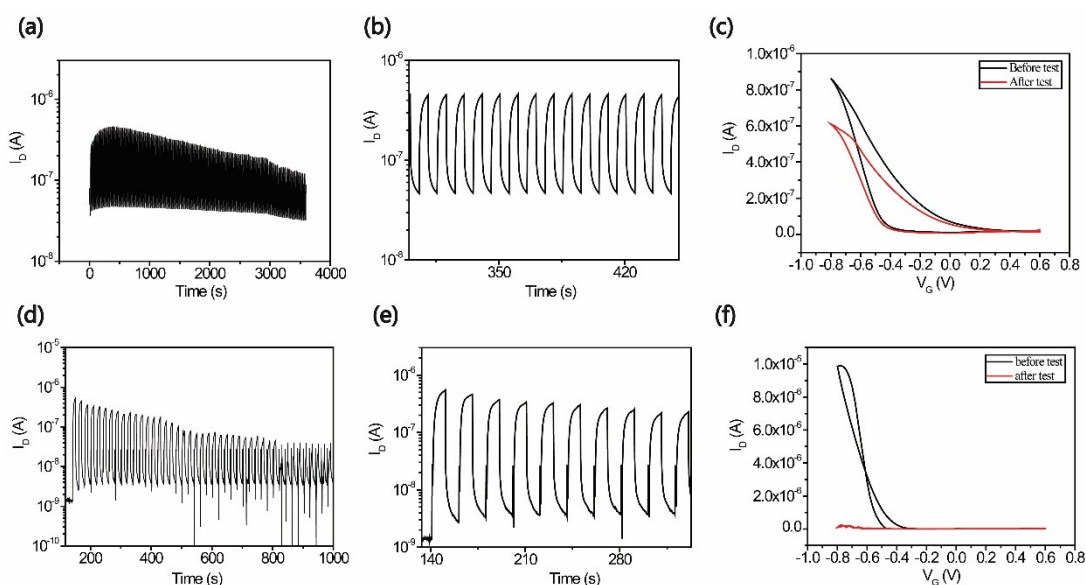


Figure S14. (a) Stability test of P1 based OEET under sequential pulsed gate voltage for 1 h with (b) an associated enlargement display from 300 s to 450 s. (V_G switches from 0.2 V to -0.4 V, pulse length = 10 s) (c) Transfer characteristics of P1 based OEET before (black) and after (red) the stability test. (d) Stability test of P3 based OEET under sequential pulsed gate voltage for 856 s with (e) an associated enlargement display from 133 s to 332 s. (f) Transfer characteristics of P3 based OEET before (black) and after (red) the stability test.

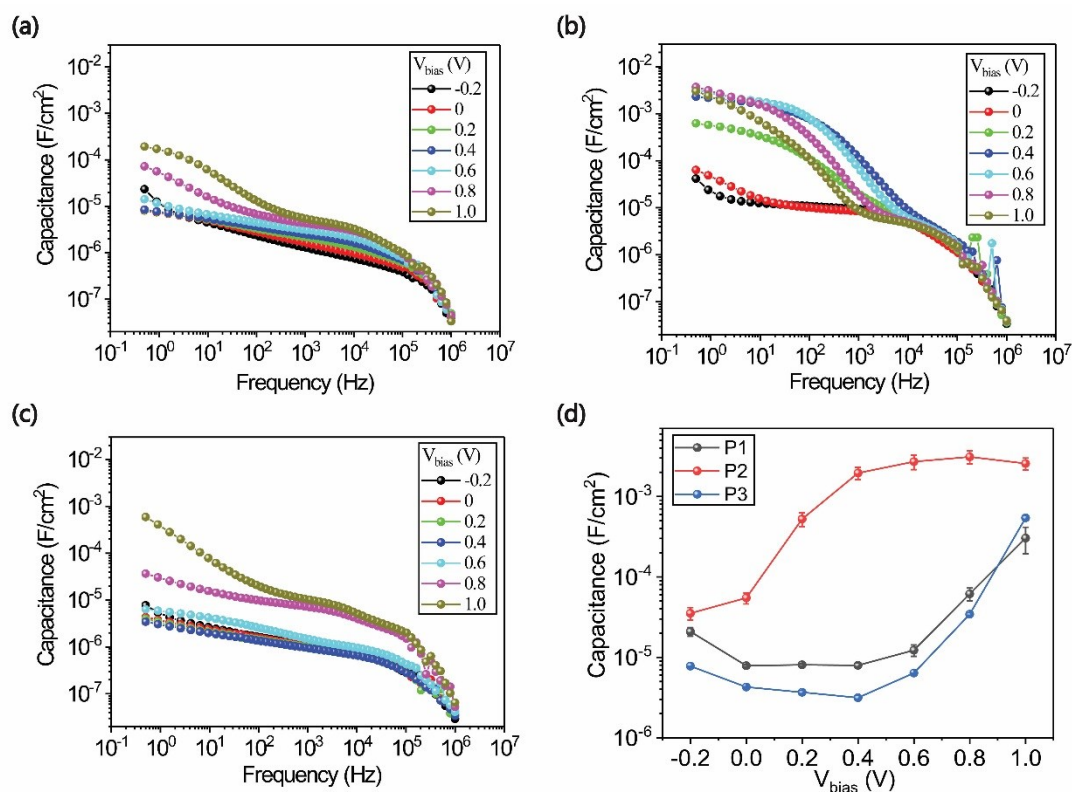


Figure S15. Effective areal capacitance from the electrical impedance spectroscopy for (a) P1, (b) P2 and (c) P3 films coated on ITO substrates. (d) Effective capacitance value at 0.5 Hz for P1 (black), P2 (red) and P3 (blue) under different bias voltage V_{bias} from -0.2 V to 1.0 V. Area of the ITO electrode: 0.2 cm².

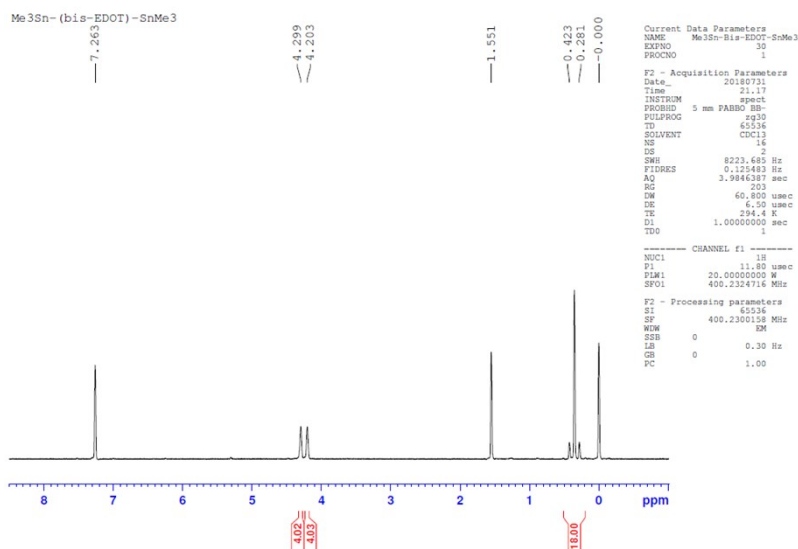


Figure S16. ¹H NMR of **8**.

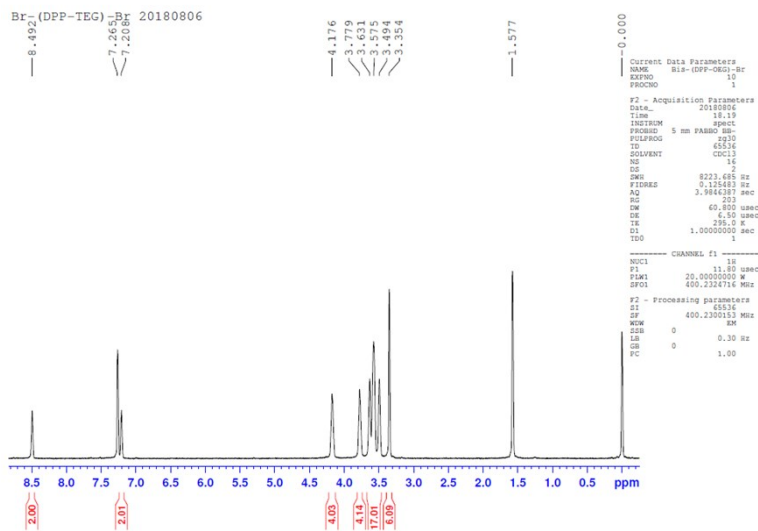


Figure S17. ¹H NMR of 6.

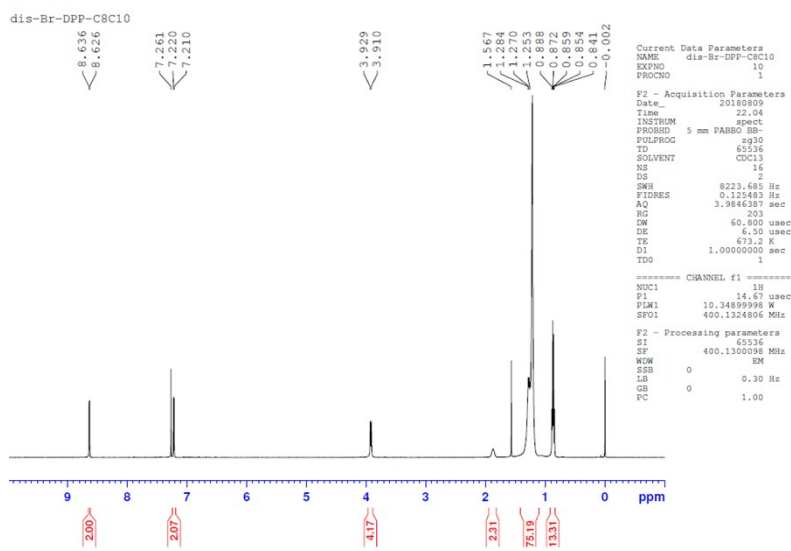


Figure S18. ¹H NMR of 3.

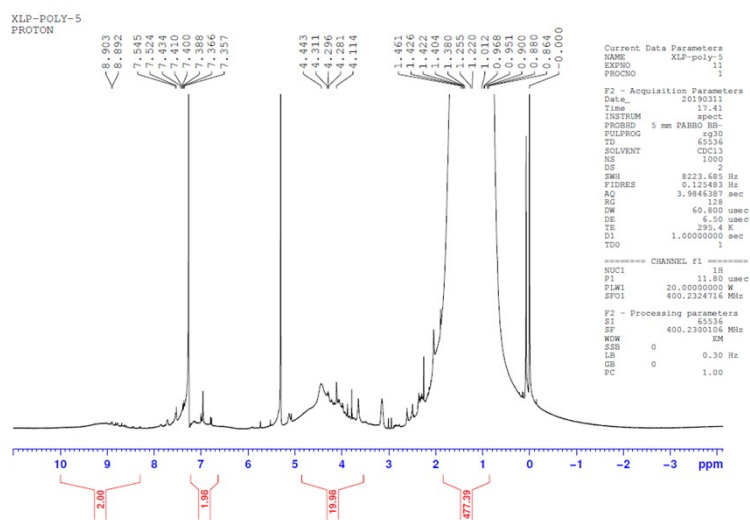


Figure S19. ¹H NMR of P1.

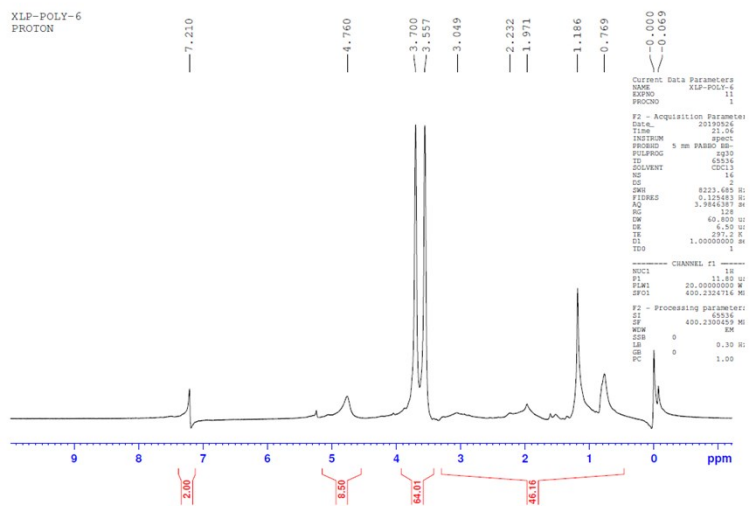


Figure S20. ¹H NMR of P2.

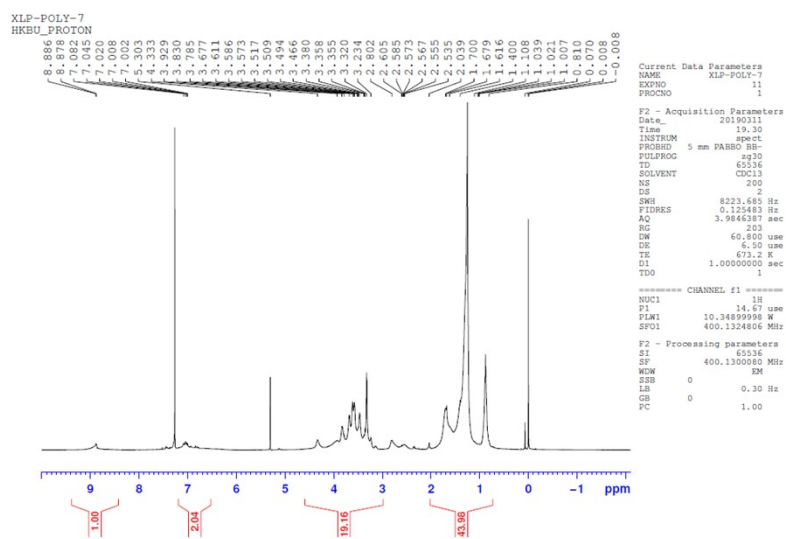


Figure S21. ¹H NMR of P3.

Table S4 Performance comparison of DPP based polymers and OECTs

| Polymer | Anions [0.1 M] | $g_m/(dW/L)$ ($S \cdot cm^{-1}$) | μ ($cm^2 \cdot v^{-1} \cdot s^{-1}$) | C^* ($F \cdot cm^{-3}$) | μC^* ($F \cdot cm^{-1} \cdot v^{-1} \cdot s^{-1}$) | Stability test duration (cycles) | Percentage retention ($I_D/I_{D,0}$) | Ref. |
|------------------------|-------------------|---------------------------------------|---|--------------------------------|--|--|--|--------------|
| p(gPyDPP- MeOT2) | Cl ⁻ | 19.5 | 0.03 | 60 | 1.8 | 120 min (3600) | 52% | [9] |
| PTDPP-DT | Cl ⁻ | 19 | 1.1 | 123 | 149 | 16 min (500) | 96% | [10] |
| p(gDPP- T2) | Cl ⁻ | 63 | 1.55 | 196 | 342 | 100 min (600) | 53% | [11] |
| P(gDTDPP- bis-EDOT) | Cl ⁻ | 3.63 | 0.087 | 189 | 16.5 | 480 min (4773) | 89.8% | This work |

References

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